MINISTÉRIO DA CIÊNCIA E TECNOLOGIA
INSTITUTO NACIONAL DE PESQUISAS ESPACIAIS

INPE-11641-PRE/7022

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Solange Tâmara da Fonseca
Maria do Carmo de Andrade Nono
Pedro José de Castro
Carlos Kuranaga
Tatiana Regina da Fonseca

Trabalho apresentado no Congresso SBPMat 2004 e submetido para publicação na Revista Ibero-Americana de Materiais – Materials Research ISSN 1516-1349

INPE
São José dos Campos
2004

THE INFLUENCE OF THE SrO_2 ON THE $Ba_2Ti_9O_{20}$ CERAMIC MICROSTRUCTURE FOR DIELECTRIC RESONATOR APPLICATIONS

Solange Tamara da Fonseca[1], Maria do Carmo de Andrade Nono[1], Pedro José de Castro[2], Carlos Kuranaga[1], and Tatiana Regina da Fonseca[1]

[1]Associated Laboratory of Sensors and Materials (LAS)

[2]Associated Plasma Laboratory (LAP)

National Institute for Space Research (INPE)

São José dos Campos, SP, Brazil

C.P. 515 - 12245-970

Abstract: This work deals with influence of strontium oxide (SrO_2) on the dielectric properties of the barium nanotitanate ($Ba_2Ti_9O_{20}$) ceramics as dielectric resonator for telecommunications applications. For these ceramics a high value both of the dielectric constant (ε) and of the quality factor (Q), as well as a low temperature coefficient of resonance frequency (τ_f) are required. The ceramics were prepared using a suitable powder mixtures, without and with SrO_2 addition. The powders were mixed, compacted by uniaxial and isostatic pressing, and sintered at 1300°C for 3 hours. The ceramics characterization was carried out using X-ray diffraction technique, and scanning electron microscopy – SEM. The dielectric parameters in microwaves were measured using a suitable microwave system. SEM analyses showed a high densification degree. The increased Q, ε and τ_f is directly proportional with the amount of SrO_2 added to the ceramics. The doped ceramic with 0.2 mol % SrO_2 presented the lowest thermal coefficient.

Keywords: dielectric resonators, barium nanotitanate ceramic, microwave characterization

Introduction

Several ceramic compositions based on barium and titanium compounds have received much attention for microwave application. Such compounds have presented a high value of both the dielectric constant (ε) and of the quality factor (Q), as well as a low temperature coefficient of resonance frequency (τ_f), small dimension, low cost, ease of assembling and compatibility with microwave integrated circuits[1-6]. This dielectric ceramic have been developed for applications in microwave telecommunications, satellite broadcasting systems and in many kinds of personal communication systems.

Microstructures of barium nanotitanate ($Ba_2Ti_9O_{20}$) dielectric resonators (DRs) doped with strontium (SrO_2) for microwave application are here studied. These DRs should fulfill the requirements of the dielectric constant as well as the quality factor high values and frequency high stability, for application as local oscillator of an INPE Communication Satellite. It has been demonstrated that the ceramic porosity affects the ε_r, the Q and the τ_f of the $Ba_2Ti_9O_{20}$ DR, as shown in Figure 1[4].

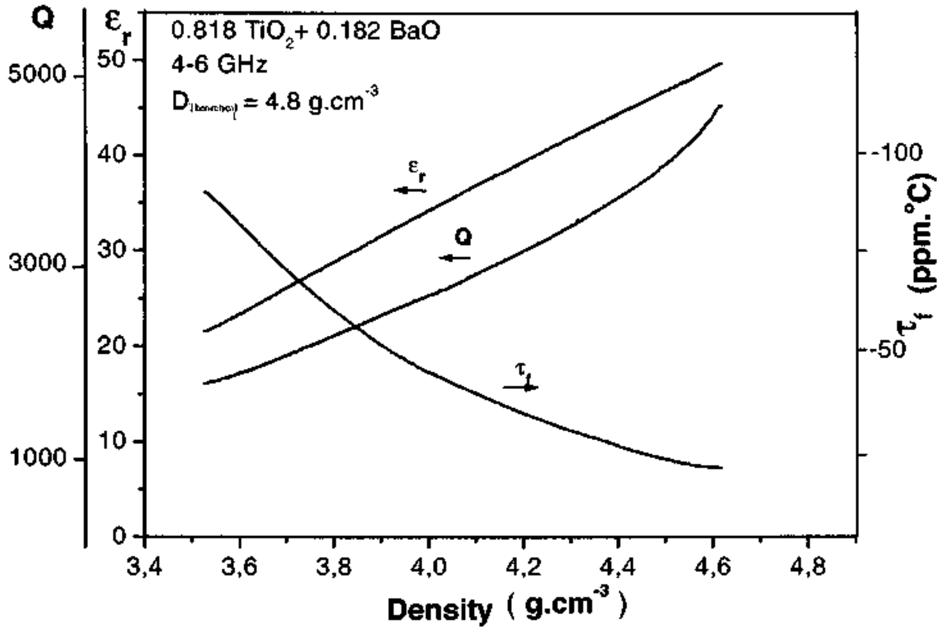


Figure 1. Effects of the ceramic density on the dielectric properties of the $Ba_2Ti_9O_{20}$ DR [4].

Experimental Procedure

Five different samples were prepared: an undoped $Ba_2Ti_9O_{20}$ specimen and other ones with 0.2, 0.4, 0.6, 0.8 and 1.0 mol % strontium contents. For solid state reaction of those ceramics, the stoichiometry, and fabrication parameters must be accurately controlled, because there are several thermodynamically stable compounds around of the desired composition of BaO–TiO_2 system. For that, the suitable 18.2 % BaO and 81.8 TiO_2 % mol composition fractions were used (Figure 2). The raw materials used for the preparation of these $Ba_2Ti_9O_{20}$ ceramics with addition of strontium were: $BaCO_3$, TiO_2 and $SrCO_3$, with purity degree more than 99 %. Since the stability region of the $Ba_2Ti_9O_{20}$ phase occurs in a narrow range of composition BaO - TiO_2 it is possible to obtain it, reacting 81.8 mol % TiO_2 and 18.2 mol % BaO (Figure 2) for temperature lower than 1300°C [4].

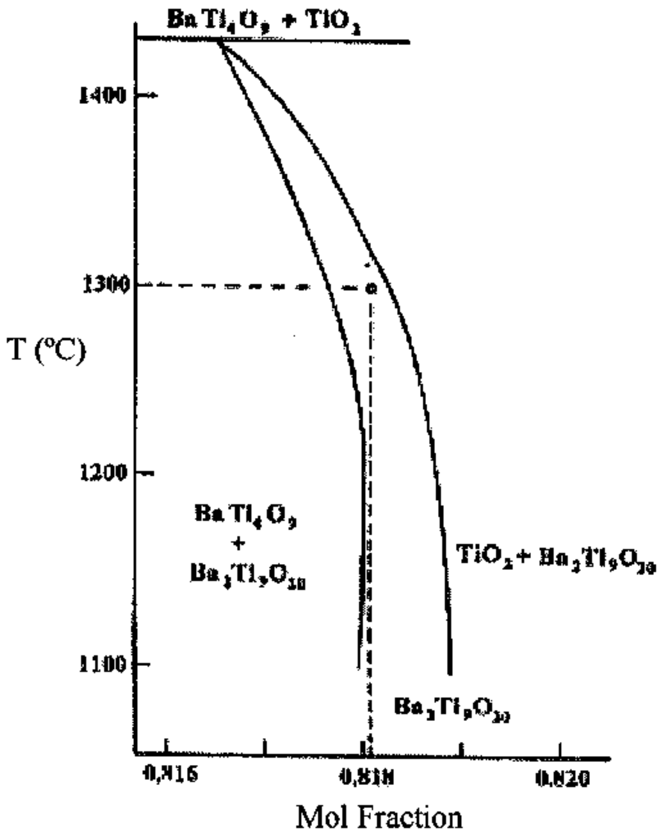


Figure 2. Equilibrium phase diagram for the TiO_2-BaO system showing the stabilization region of the $Ba_2Ti_9O_{20}$ phase [4].

The manufacturing process of these DRs requires the suitable temperature both for the formation and the stabilization of the $Ba_2Ti_9O_{20}$ phase and for the sintering and densification of the ceramic.

Cylindrical samples with a preestablished relationship of H/D = (0.40 ± 0.02) (where H is height and D the diameter) are produced in order to operate at the desired frequency range 5.7-6.8 GHz.

Once defined the composition, the powders of $BaCO_3$ and TiO_2 with and without addition of $SrCO_3$ were mixed in ethyl alcohol using magnetic agitators for 30 minutes, this way provides a sample free from impurities.

The powders were mixed and dried using a rotative evaporator at vacuum. This process aided the total homogeneity and the break of the formed agglomerates, thus, minimizing the formation of kind of particles. The powder was compacted by a uniaxial (40 MPa) and isostatic (300 MPa) pressings, producing cylindrical test bodies.

Finally, they were synthesized and sintered at 1300°C for 3 hours. Afterwards, the ceramic crystallographic phases were characterized by X-rays diffraction. The fracture surfaces were observed by SEM.

The dielectric parameters were measured at microwave frequencies: resonance frequency (f), dielectric constant (ε), thermal coefficient (τ_f) and, the unloaded quality factor Q_o, which in its turn corresponds to the Q factor due to dielectric losses. These characteristics were measured using a test box made from copper covered by gold. The testing device consists of a cylindrical dielectric radius a and height H, placed between two parallel conducting plates, in the case of resonant frequency measurement [3, 5]. This configuration allows that DR can operate at the electromagnetic mode TE_{011}. From the experimental value of the resonant frequency we can determine the dielectric constant by an electromagnetic field equation relating the resonant frequency, the dielectric constant and the resonator dimensions. The calculation was made running the software "Mathematica" in a microcomputer [3,5]. As for the quality factor, providing the test box dimensions are at least three times the size of the dielectric resonator and inserting the resonator between low-loss teflon spacers, then the metallic losses can be ignored. The microwave measurements were performed according to the test setup shown in Figure 3 [3]. The DR was excited by means of an electric probe with optimum coupling. Another electric probe is used as a receiving device to detect the sign radiated by the resonator. The measured factor quality is the unloaded Q (Q_o), based on bandwidth at the half-power frequencies on the detected frequency spectrum.

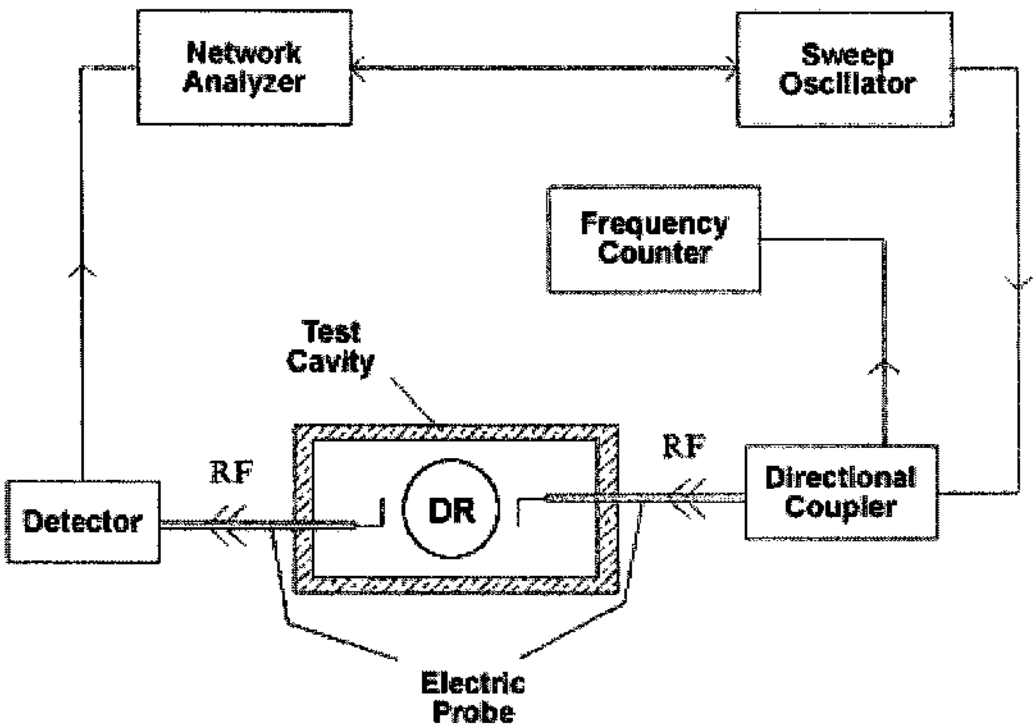


Figure 3. Experimental setup for measuring the microwave parameters [3].

The resonance frequency variation with temperature is valuated by the thermal coefficient and represented by symbol (τ_f). This parameter is measured using the same setup showed on the Figure 3, however it was put inside of a thermal chamber. This chamber operated with air flow under forced convection and with nitrogen gas in order to keep the atmosphere as dry as possible. The resonance frequency measured around 5.12 GHz on the temperature range from -20°C to +50°C.

The thermal coefficient was determined using the following equation:\

$$\tau_f = \frac{1}{f_i}\frac{\Delta f}{\Delta t} \quad [\text{ppm/°C}] \qquad (1)$$

where Δf is difference between final and initial frequencies, Δt is difference between final and initial temperatures and f_i is the resonance frequency at the beginning of process. The abbreviation "ppm" (parts for million) is often used to denote the term 10^{-6}.

It is known that the thermal coefficient depends on the expansion coefficient of the dielectric material, the expansion coefficient of the surrounding material (copper) and the thermal coefficient of the dielectric constant variation with temperature.

Results and Discussion

For the doped ceramics from 0.2 to 0.8 mol % SrO_2 only the phase $Ba_2Ti_9O_{20}$ was identified (Figure 4), therefore a small quantity of SrO_2 did not affect the formation of the other phases. In particular, the doped ceramic with 1.0 mol % SrO_2 contains the $BaTi_4O_9$ phase formation that may have influenced negatively in some microwave parameters.

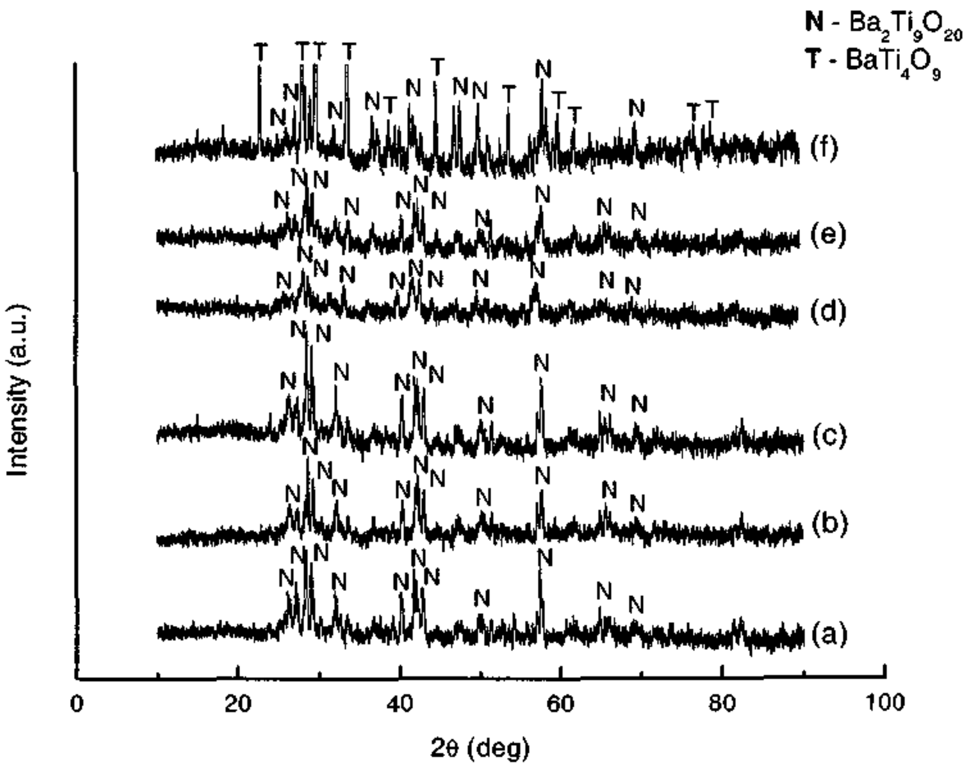
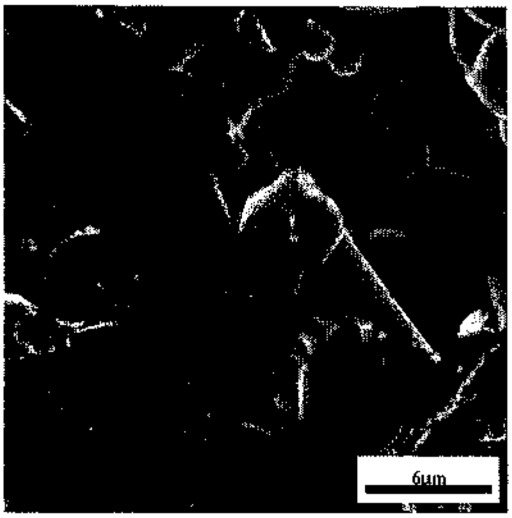
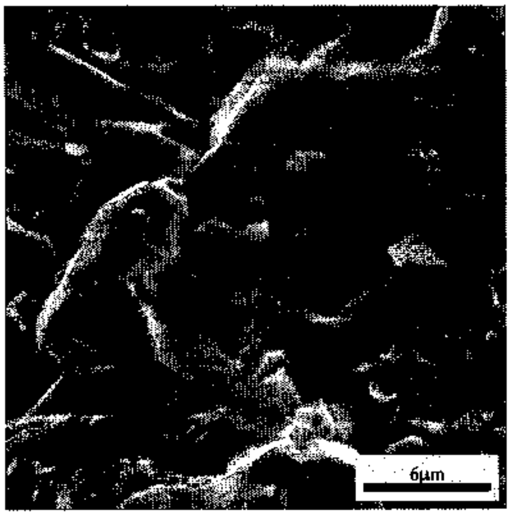


Figure 4. X-rays diffraction patterns for the $Ba_2Ti_9O_{20}$ ceramics: (a) without SrO_2 addition, (b) with 0.2 mol % SrO_2, (c) with 0.4 mol % SrO_2, (d) with 0.6 mol % SrO_2, (e) with 0.8 mol % SrO_2 and (f) with 1.0 mol % SrO_2.

The scanning electronic microscopy (SEM) images of the ceramic fracture surfaces showed a high densification degree of the microstructure and the presence of few pores (Figure 5)

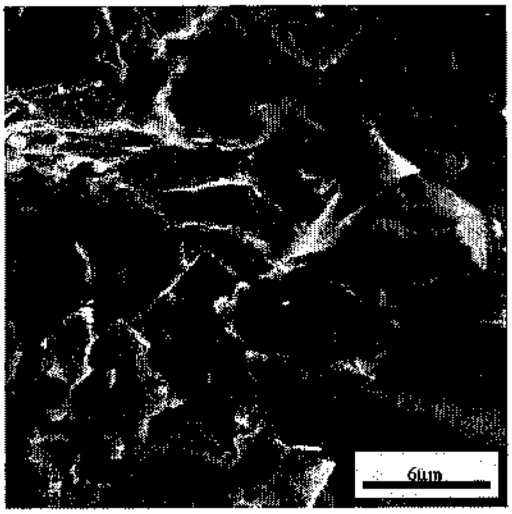


(a)

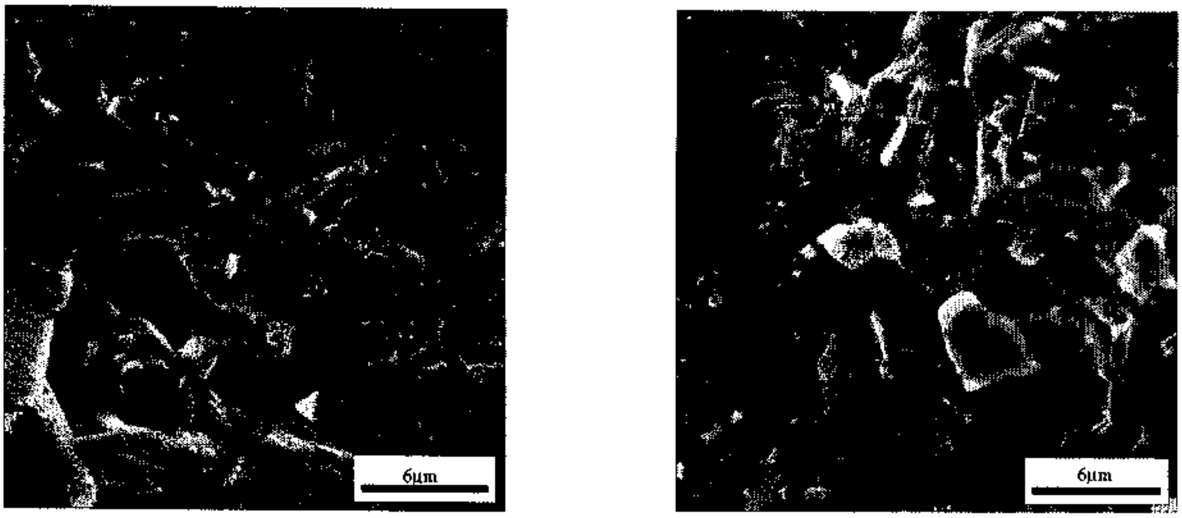


(b)

(c)



(d)



(e) (f)

Figure 5. SEM micrographs of $Ba_2Ti_9O_{20}$ ceramic fracture surfaces: (a) without SrO_2 addition, (b) with 0.2 mol % SrO_2, (c) with 0.4 mol % SrO_2, (d) with 0.6 mol % SrO_2, (e) with 0.8 mol % SrO_2 and (f) with 1.0 mol % SrO_2.

General ceramics presented a high degree of densification not being observed the influence of strontium addition.

The measuring characteristics at microwave frequencies for investigated DRs are presented in the Table 1. The thermal coefficient was valuated by the average value from measured frequencies for every 5°C step. The dielectric parameters were affected with SrO_2 increase: both the quality factor due to dielectric losses and the dielectric constant became higher in the most part of the samples. In particular, the doped ceramic with 1.0 mol % SrO_2 has decreased its Q factor, probably it was caused by influence of the $BaTi_4O_9$ phase, which has lower quality factor than $Ba_2Ti_9O_{20}$ one. The thermal coefficients have also become higher. The thermal coefficients presented relatively good results, thus providing frequency stability for dielectric resonators, in this sense the ceramic doped with 0.2 mol % SrO_2 got the best result.

Table 1 – The measuring results at microwave frequencies for investigated RDs

SrO₂ *%*	***H ± 0.01*** *(mm)*	***a ± 0.01*** *(mm)*	$f \pm 1.5x10^{-3} \rightarrow \epsilon_r \pm 0.3$ *(GHz)*	Q_0 *(*)*	***τ (**)*** *(ppm/°C)*
0.0	**4.53**	**5.69**	**7.6293 → 30.8**	**3.488**	**6.2**
0.2	**4.45**	**5.65**	**7.5525 → 32.3**	**3.000**	**4.6**
0.4	**4.37**	**5.61**	**7.4525 → 33.3**	**3.640**	**-**
0.6	**4.38**	**5.54**	**7.3450 → 35.4**	**3.924**	**8.0**
0.8	**4.38**	**5.56**	**7.3751 → 35.0**	**4.715**	**-**
1.0	**4.39**	**5.51**	**7.3356 → 35.5**	**4.230**	**12.2**

(*) Q factor measured around 5.0 GHz
(**) Thermal coefficient measured around 5.12 GHz on the temperature range from -20°C to +50°C

Conclusions

The synthesizing and simultaneous sintering procedures make possible an increase of the suitable densification degree at lower temperature in comparison to those ceramics obtained by other authors.

Strontium addition has aided the complete $Ba_2Ti_9O_{20}$ phase stabilization for dielectric resonators, and so the Q factor has. As for the measured thermal coefficients, DRs with 0.0 and 0.2 mol % SrO_2 have got relatively goods results. As for the ceramics having not so low thermal coefficient, their behavior probably was not related to the ceramic porosity, but to the fact that the increase of strontium amount provides a positive thermal coefficient portion and you should supply a certain compound with negative thermal coefficient in order to compensate it.

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